Layered Ultrathin PbI_2 Single Crystal for High Sensitivity Photodetector

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Designing of mineralizer:

Considering that the (001) plane of PbI_2 tends to be terminated by I^-, in order to restrict the growth rate along the [001] direction and enhance the two dimensional growth of the single crystals, the basic growth units should be negatively charged. KI was usually used as mineralizer for growing iodide single crystals, which can generate negative charged complex ions. However, for our system, excessive KI may lead to the formation of $KPbI_3$. Therefore, we choose KNO_3 (1 M) as the mineralizer. The schematic illustration of the growth habit of PbI_2 in presence of KNO_3 mineralizer is shown in Figure S1. As shown in Figure S1, adding high-concentration NO_3^- ions to the solution may lead to the formation of the intermediate complex $[PbI_2(NO_3)]^{2-}$ and thus increased the solubility of PbI_2. More importantly, the basic growth units in such a growth environment should be the negatively charged $[PbI_2(NO_3)]^{2-}$ ions, which are less likely to be absorbed on the I-terminated (001) plane. Therefore, the growth rate along the [001] direction is restricted, and the continuous two-dimensional growth can be achieved.

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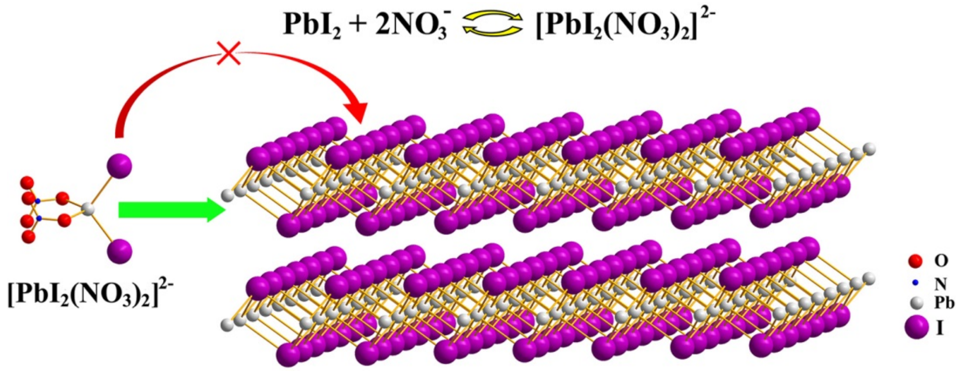

Figure S1. Growth habits of PbI_2 controlled by the KNO_3 mineralizer.

Electric characteristics of PbI_2 with different metal contacts:

To evaluate the influence of different metal contacts on the electric characteristics of PbI_2, we have deposited different metals (Au, Cu, Ag, 50 nm thick) on the ultrathin PbI_2 single crystals (300 nm thick using mechanical exfoliation from the thin PbI_2 single crystals) through a Cu grid shadow mask to form two electrodes with a gap of 20 μm. The current-voltage (*I*-*V*) characteristics of the devices were measured in dark and under the illumination of a 407 nm laser diode. The dark current and light current of the devices using different metal contacts at 5 V bias were shown in Figure S2. It can be seen that under our experimental conditions, with the increasing of the work function of metal contacts, the dark current of the devices decreased dramatically, while the light current were not significantly changed. Thus, Au contacts can effectively improve the ON/OFF ratio of the PbI_2 photodetectors. Here, it should be noted that decreasing the thickness of PbI_2 single crystal is also an effective way to reduce the dark current of the lateral M-S-M photodetectors.

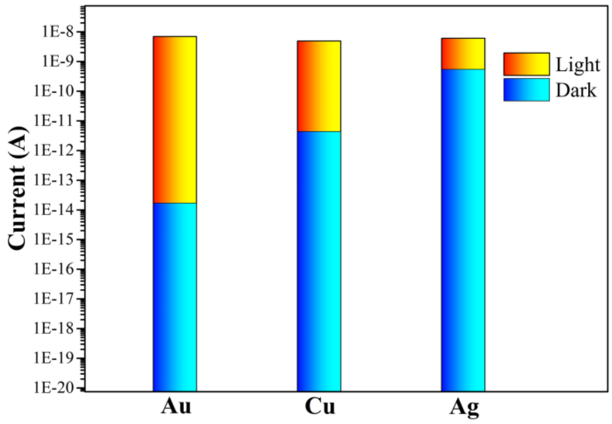

Figure S2. Dark current vs. light current of the devices using different metal contacts at 5 V bias.

Furthermore, to exhibit the contact behavior of Au on PbI_2, we also fabricate a device on a 2 μm thick PbI_2 single crystal with one 2 × 2 mm Au contact on the top and another on its back. The *I-V* measurements (shown in Figure S3) showed that the Au contacts exhibit a non-Ohmic rectifying behavior, which may be responsible for the suppressed dark current.

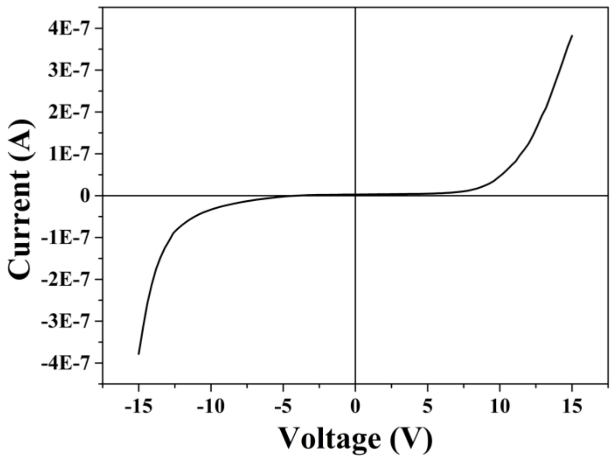

Figure S3. *I-V* curves of Au metal contacts on 2 μm thick PbI_2 single crystal in a vertical configuration.